## **Description of Additional Supplementary Files**

File Name: Supplementary Movie 1

Description: Supplementary Movie 1 for the peeling process. In Supplementary Movie 1, we use a spoon to peel off the laminated, pliable, free-standing COF membrane (FS-COM-1) collected from the organic-organic interface. What we want to express in the video is that the layer-by-layer nanosheets can be peeled off easily, even with a spoon.

File Name: Supplementary Movie 2

Description: Supplementary Movie 2 for the light-driven progress. In Supplementary Movie 2, what we want to express in the video is the low density of the membrane. We tried to hit the COF membrane (~ 0.7 mg) with a laser and to see if it could be driven (The power of the laser pointer is 3 mW). To prevent the influence of air flow, the experiment was carried out in a closed balance. And in the experiment, COF nanosheet and capillary tube were linked by spider silk. Delightedly, the COF membrane can do be driven by the red laser. A video about the light-driven process is attached in the Supplementary Movie 2.